

Claims

1. A homogeneous process for the hydrogenation of dicarboxylic acid and/or derivative thereof with an amine in the presence of a catalyst comprising:
 - (a) ruthenium or osmium; and
 - (b) an organic phosphine;and wherein the hydrogenation is carried out in the presence of water.
2. A homogeneous process according to Claim 1 wherein the water is present in at least 1% by weight.
3. A homogeneous process according to Claim 1 or 2 wherein the dicarboxylic acid and/or derivative thereof is selected from one or more of dicarboxylic acids, polycarboxylic acids, anhydrides, monoesters or diesters of dicarboxylic acids, monoamides or diamides of dicarboxylic acids, salts, such as amine salts, of dicarboxylic acids or mixtures thereof.
4. A homogeneous process according to any one of Claims 1 to 3 wherein the dicarboxylic acid and/or derivative thereof is a C₄ to C₁₂ dicarboxylic acid.
5. A homogeneous process according to any one of Claims 1 to 4 wherein the dicarboxylic acid and/or derivative thereof is saturated or unsaturated.
6. A homogeneous process according to any one of Claims 1 to 5 wherein the dicarboxylic acid and/or derivative thereof is selected from maleic acid, fumaric acid, succinic acid, maleic anhydride, adipic acid, and succinic anhydride.

7. A homogeneous process according to any one of Claims 1 to 6 wherein the amine is $\text{NR}^1\text{R}^2\text{R}^3$ where R^1 , R^2 and R^3 are each selected from hydrogen or alkyl.
8. A homogeneous process according to Claim 7 wherein the alkyl group is a C_1 to C_6 alkyl group.
9. A homogeneous process according to Claim 8 wherein the alkyl group is methyl, ethyl or propyl.
10. A homogeneous process according to any one of Claims 1 to 9 wherein the amine is mixed with the dicarboxylic acid and/or derivatives thereof prior to undergoing the process.
11. A homogeneous process according to any one of Claims 1 to 10 wherein the dicarboxylic acid and/or derivative thereof is converted to the amide by reaction with the amine prior to commencement of the hydrogenation.
12. A homogeneous process according to any one of Claims 1 to 11 wherein the water is present as the solvent for the reaction.
13. A homogeneous process according to any one of Claims 1 to 11 wherein one or both of the reactants or the product of the reaction are the solvent.
14. A homogeneous process according to any one of Claims 1 to 13 wherein a solvent is used and water is present as an additive in the solvent.
15. A homogeneous process according to Claim 14 wherein the solvent is selected from tetraethyleneglycol dimethyl ether, N-methyl pyrrolidone, diethyl ether, ethyleneglycol

dimethylether, dioxane, 2-propanol, 2-butanol, secondary alcohols, tertiary alcohols, lactams and N-methyl caprolactam.

16. A homogeneous process according to any one of Claims 1 to 11 wherein the water is produced in situ as a by-product of the hydrogenation reaction.
17. A homogeneous process according to any one of Claims 1 to 16 wherein the catalyst comprises ruthenium and the ruthenium is provided as a ruthenium compound.
18. A homogeneous process according to Claim 17 wherein the ruthenium compound is a compound selected from nitrates, sulphates, carboxylates, beta diketones, or carbonyls.
19. A homogeneous process according to any one of Claims 1 to 18 wherein the ruthenium is present in an amount of from 0.0001 to 5 mol, preferably 0.005 to 1 mol, as ruthenium per liter of reaction solution.
20. A homogeneous process according to any one of Claims 1 to 19 wherein the phosphine is a tridentate phosphine.
21. A homogeneous process according to any one of Claims 1 to 19 wherein the phosphine is selected from trialkylphosphines, dialkylphosphines, monoalkylphosphines, triarylphosphines, diarylphosphine, monoarylphosphines, diarylmonoalkyl phosphines and dialkylmonoaryl phosphines.
22. A homogeneous process according to Claim 21 wherein the phosphine is selected from tris-1,1,1-(diphenylphosphinomethyl)methane, tris-1,1,1-(diphenylphosphinomethyl)-ethane, tris-1,1,1-(diphenylphosphinomethyl)propane, tris-1,1,1-(diphenylphosphino-methyl)butane, tris-1,1,1-(diphenylphosphinomethyl)2,2dimethylpropane, tris-1,3,5-

(diphenylphosphino-methyl)cyclohexane, tris-1,1,1-(dicyclohexylphosphinomethyl)ethane, tris-1,1,1-(dimethylphosphinomethyl)ethane, tris-1,1,1-(diethylphosphinomethyl)ethane, 1,5,9-triethyl-1,5,9-triphosphacyclododecane, 1,5,9-triphenyl-1,5,9-triphosphacyclododecane, bis(2-diphenylphosphinoethyl)phenylphosphine, bis-1,2-(diphenyl phosphino)ethane, bis-1,3-(diphenyl phosphino)propane, bis-1,4-(diphenyl phosphino)butane, bis-1,2-(dimethyl phosphino)ethane, bis-1,3-(diethyl phosphino)propane, bis-1,4-(dicyclohexyl phosphino)butane, tricyclohexylphosphine, trioctyl phosphine, trimethyl phosphine, tripyridyl phosphine and triphenylphosphine.

23. A homogeneous process according to Claim 21 wherein the phosphine is selected from tris-1,1,1-(diarylphosphinomethyl)alkane and tris-1,1,1-(dialkylphosphinomethyl)alkane.
24. A homogeneous process according to any one of Claims 1 to 23 wherein the catalyst is preformed.
25. A homogeneous process according to any one of Claims 1 to 24 wherein the phosphine compound is present in an amount of from 0.0001 to 5 mol as phosphine per liter of reaction solution.
26. A homogeneous process according to any one of Claims 1 to 25 wherein the hydrogenation is carried out at temperatures in the region of from about 190°C to about 300°C.
27. A homogeneous process according to any one of Claims 1 to 26 wherein the reaction pressure is from about 500 psig to about 2000 psig.
28. A homogeneous process according to any one of Claims 1 to 27 wherein the ratio of amine to acid or derivative thereof is from about 0.5:1 to about 100:1.

29. A homogeneous process according to any one of Claims 1 to 28 wherein the ratio of amine to acid or derivative thereof is from about 0.9:1 to about 2.0:1.
30. A homogeneous process according to any one of Claims 1 to 29 wherein the product is a 2-pyrrolidone or an N-alkylated version thereof and the dicarboxylic acid or derivative thereof is maleic acid, maleic anhydride, succinic acid or succinic anhydride.
31. A homogeneous process according to any one of Claims 1 to 29 wherein the product is caprolactam and the dicarboxylic acid or derivative thereof is adipic acid.
32. A homogeneous process according to any one of Claims 1 to 31 wherein the catalyst is regenerated in the presence of the water and hydrogen.